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Project Summary

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Coal Gasifier Parameters Influencing Environmental Pollutant Production

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A series of fixed-bed coal gasification and pyrolysis tests have been performed in a laboratory-scale reactor of 6.6 cm inside diameter. Chemical analyses were conducted on the product gas, aqueous condensate, tar, and solid residue from the tests. The effects of process variables, such as feed mode, catalytic treatment, pressure, mesh size, and coal type, by-product and pollutant yields are described. The production of gaseous sulfur compounds, benzene and derivatives (BTX), phenolics (phenol, cresols, and xylenols), and tar has been measured, and the fate of trace elements such as arsenic, selenium, and lead has been determined. By-product production associated with the pyrolysis phase of gasification has been investigated, with emphasis on the effects of particle size, residence time, and atmosphere.

Experiments with six U.S. coals ranging from lignite to anthracite are described. Continuous operation resulted in greatly reduced tar and phenolic production. When the entire mass of coal was introduced at the beginning of the test, effluent stream compositions more closely approached those of larger scale, fixed-bed gasifiers. Initial work on this project was reported in EPA-600/7-78-171 (NTIS PB287916); three more recent reports were EPA-600/7-79-200 through EPA-600/7-79-202 (NTIS PB80-182769, 80-104656, and 81-114308).

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Technology for the conversion of coal to gaseous fuel has been available for many years. Renewed interest in coal gasification processes, motivated by greatly increased petroleum prices and anticipated shortages of oil and natural gas, has stimulated considerable research into process improvements. Gaseous fuels are inherently easier to transport and distribute than coal. With increasing constraints on the discharge of oxides of sulfur and nitrogen, gaseous fuels may require less expensive pollution control systems than coal and oil. Additionally, indirect liquefaction of coal via catalytic reaction of coal-derived gas to produce oil substitutes is approaching economic feasibility in the U.S.

The increasing likelihood of large-scale commercial development of coal gasification in the U.S. has generated concern over the pollutants produced in these processes. The Research Triangle Institute has performed a 5-year study directed toward quantifying the production of by-products and pollutants from coal gasification processes and investigating the influence of processing

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conditions on these materials. The project was sponsored by the Industrial Environmental Research Laboratory of the U.S. Environmental Protection Agency at Research Triangle Park, NC.

The experiments were conducted in a laboratory-scale coal gasification system designed and constructed at RTI. The reactor is a 100-cm length of nominal 3-in. Schedule 160, type 310 stainless steel pipe, fitted with ring joint flanges at both ends. A flow distributor, a porous ceramic disk mounted on a stainless steel plenum chamber, is inserted into the reactor to a height of approximately 30 cm from the bottom reactor flange. The flow distributor supports the coal bed. The reactor is enclosed in a vertical tube furnace with three independently controlled heating zones. Superheated steam is generated by pumping deionized water with a positive-displacement, packed, plunger-type metering pump through stainless steel tubing enclosed in a series of three electrically heated furnaces. Nitrogen, oxygen, or air from standard compressed gas cylinders are mixed with the superheated steam and enter the reactor through a tube connecting the bottom flange and the flow distributor.

Coal was added to the reactor from the top. Usually coal from a pressurized feed hopper was added batchwise through a pneumatically operated ball valve. In some cases a rotating-disk coal feeder was used to continuously supply coal to the reactor. Gases flowed out of the top of the reactor through a tube in the upper flange into a water-jacketed condensate trap. A two-phase mixture collected in this trap: a tar phase and an aqueous condensate phase made up of moisture from the coal and unconverted reactant steam. The condensate trap was emptied periodically through a pressure lock arrangement using two remotely operated ball valves. The gas flowed from the condensate trap through an adjustable back pressure regulator where the pressure was reduced to slightly above atmospheric. The gas then entered a sampling manifold with provisions for discrete sampling in glass bulbs, resin adsorption, and an impinger. Finally, the gas passed through a dry test meter and was vented.

The reactor temperature was determined using a multipoint thermowell containing up to 12 chromel-alumel thermocouples. This thermowell was inserted through the upper flange of the reactor to a point slightly above the flow distributor. The thermocouples initially were connected to digital displays. For the more recent tests, the thermocouples were also connected to analog-to-digital converters, with readings automatically stored on magnetic disks. Additional thermocouples were used to measure the temperature of the inlet steam and condensate trap. The pressure at the steam-air inlet and at the exit of the condensate trap was measured both by transducers and by bourdon tube gauges. The transducer outputs were displayed digitally and stored.

A variety of fuels have been gasified in the laboratory system. This report deals with experiments made with the following fuels:

1. North Dakota Beulah/Zap lignite.
2. Montana Rosebud/McKay subbituminous coal.
3. Wyoming Smith/Roland subbituminous coal.
4. Illinois No. 6 bituminous coal.
5. West Kentucky No. 9 bituminous coal.
6. Pennsylvania Bottom Red Ash anthracite coal.

Conclusions

Operation of the laboratory gasifier in the continuous mode resulted in a higher ratio of CO_2 to CO, and a greater yield of benzene, toluene, and xylenes (BTX) than operation in the semicontinuous mode. The continuous gasification mode also generated less tar and less phenolic material than the semicontinuous mode, and the tar that was produced in continuous testing was lower in tar acids than that produced in semicontinuous testing. The tar produced in semicontinuous testing was quite similar to that produced in larger scale, intermittently fed, fixed-bed gasifiers in both composition and yield.

Gasification of Illinois No. 6 coal that had been treated with alkali metal compounds indicated that the catalytic treatment did not increase reaction rates, lower the reaction temperatures, or decrease caking and swelling tendencies in the laboratory reactor. Treatment with K_2CO_3 reduced H_2S production and increased sulfur and mercury retention in the solid residue but had no effect on nitrogen retention. Treatment with NaOH at a much lower level reduced tar production but did not change the elemental composition of the tar; treatment with NaOH did not affect nitrogen or sulfur retention but did increase the retention of mercury in the solid residue.

When the particle size of the coal fed to the gasifier was doubled, the product gas was higher in CO relative to CO_2. For Illinois No. 6 coal, larger particles produced less BTX and a tar that was higher in acids. Doubling the particle size of North Dakota lignite increased the tar yield.

Lower pressure gasification produced more CO relative to CO_2 and tar with a decreased polynuclear aromatic content. With Illinois No 6 coal, lower pressure operation resulted in a product gas higher in CO relative to hydrogen and a greatly increased tar yield.

The effect of rank on by-product production is shown in Figure 1. Both tar and BTX yields increased with rank from lignite to bituminous. Anthracite coal produced no tar and very little BTX. Phenol, cresols, and xylenols (PCX) production generally decreased with increasing coal rank. Lignite and subbituminous coal produced tar with a higher proportion of acids and a lower proportion of bases than the tar from bituminous coals. Consistent with behavior in combustion processes, more sulfur was retained in the solid residue from lignite and subbituminous coals than in the solid residue from bituminous coal. Nitrogen retention in gasification residue did not vary consistently with rank.

In tests with Illinois No. 6 coal, arsenic volatilization was strongly correlated with carbon and sulfur conversion. These relationships are shown in Figure 2. Selenium volatilization was significantly correlated with carbon conversion but not with sulfur conversion. Mercury was completely volatilized in all tests for which data is available except for tests where the feed coal was treated with alkali metal compounds; treatment with NaOH did, however, increase the volatility of beryllium.

Pyrolysis experiments in an inert atmosphere showed that particle size had no effect on BTX production or rate of devolatilization, but did influence drying time. When Wyoming subbituminous coal was heated in a hydrogen/CO atmosphere, initial H_2S production equaled that of pyrolysis in a nitrogen atmosphere. Additional H_2S was produced after pyrolysis was complete when the surrounding atmosphere contained hydrogen. At no stage of the pyrolysis experiment was H_2S in equilibrium with carbonyl sulfide; it was concluded that H_2S evolved from the

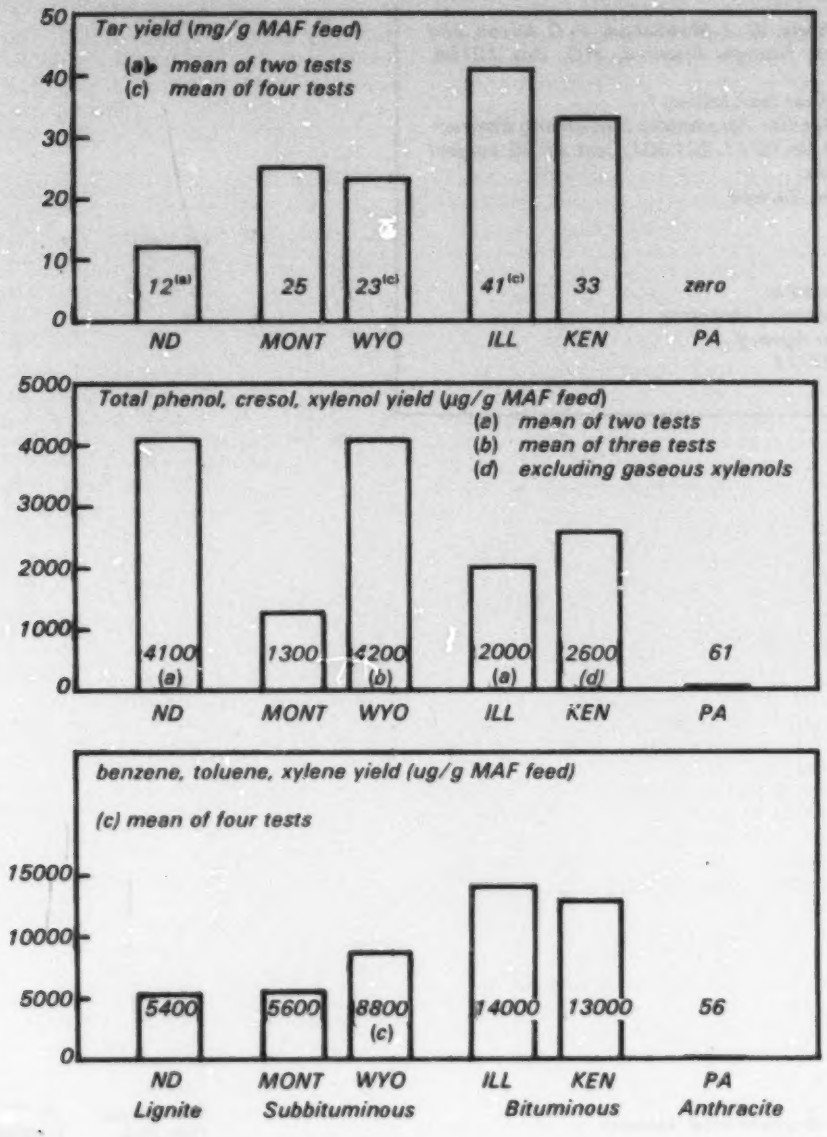

Figure 1. Effect of rank on byproduct production from airblown, fixed bed, semi-continuous gasification of 8x16 mesh coal at 200 psi.

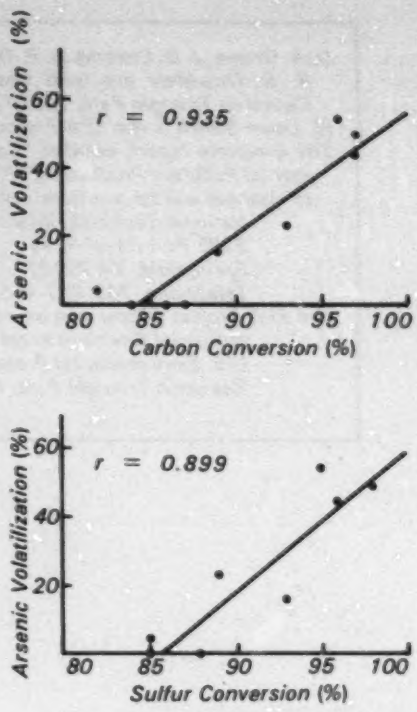

Figure 2. Correlation of arsenic volatilizatoin with A. carbon conversion and B. sulfur conversion. Gasification tests with Illinois No. 6 coal.

coal and that its hydrolysis was kinetically limited.

Recommendations

The method in which coal is fed to the gasifier was responsible for a profound change in by-product production in fixed-bed, air-blown gasification. Specifically, tar and phenolic compounds were reduced when coal was fed continuously, yet batch coal feed resulted in by-product yields which closely approximated those of larger scale intermittently fed gasifiers. If larger scale, fixed-bed gasifiers were fed smaller batches of coal at more frequent intervals, reduced pollutant yields might result.

Treating caking bituminous coal with K_2CO_3 increased the amount of sulfur retained in the solid residue, but did not affect the caking tendency of the coal. The use of alkali metals as a means of reducing gaseous H_2S is potentially valuable. It is unlikely, however, that alkali metal treatment will allow caking coals to be gasified in fluid-bed gasifiers without pretreatment.

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The complete report, entitled "Coal Gasifier Parameters Influencing Environmental Pollutant Production," (Order No. PB 81-221 301; Cost: $9.50, subject to change) will be available only from:
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